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(54) PYRIMIDIN-4-ONE DERIVATIVES AS PESTICIDE

PYRIMIDIN-4-ON DERIVATE ALS PESTIZIDESMITTEL

DERIVES DE PYRIMIDINE-4-ONE UTILISES COMME PESTICIDE

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Remarks:
The file contains technical information submitted after the application was filed and not included in this specification

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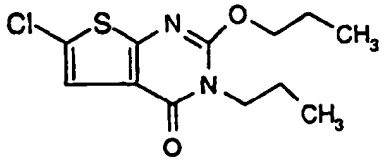
Description

[0001] The present invention relates to a novel pyrimidin-4-one derivative of formula I, which has pesticidal activity, in particular fungicidal activity,



I ,

6-chloro-2-propoxy-3-propyl-3H-thieno[2.3-d]pyrimidin-4-one.

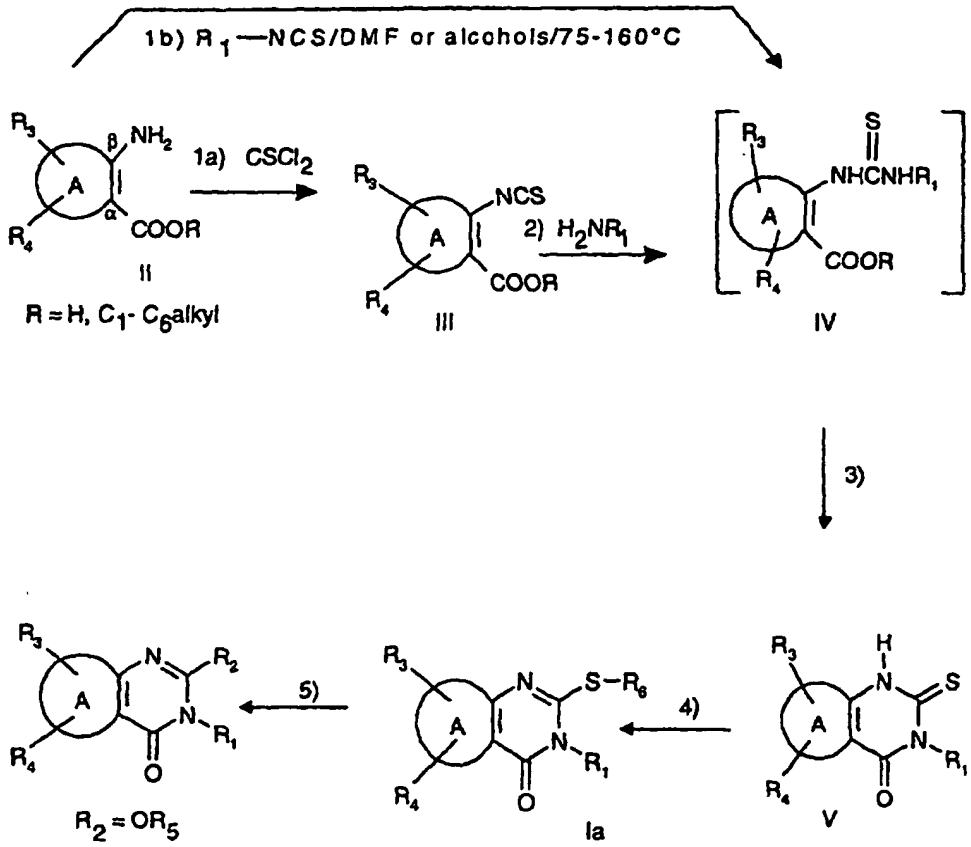
[0002] The invention also relates to the preparation of this compound, to agrochemical compositions comprising as active ingredient at least this compound, as well as to the use of the active ingredients or compositions for pest control, in particular as fungicides, in agriculture and horticulture.

[0003] The compound I and, optionally, its tautomers may be obtained in the form of their salts. Because the compound I has at least one basic centre he can, for example, form acid addition salts. Said acid addition salts are, for example, formed with mineral acids, typically sulfuric acid, a phosphoric acid or a hydrogen halide, with organic carboxylic acids, typically acetic acid, oxalic acid, malonic acid, maleic acid, fumaric acid or phthalic acid, with hydroxycarboxylic acids, typically ascorbic acid, lactic acid, malic acid, tartaric acid or citric acid, or with benzoic acid, or with organic sulfonic acids, typically methane acid or p-toluenesulfonic acid.

[0004] Within the scope of this invention, agrochemically acceptable salts are preferred.

[0005] The compound of formula I can be prepared as follows:

Scheme 1



R_1=n-propyl, R_5=n-propyl, R_3=chloro, R_4=hydrogen, A=[2.3-d]thienyl

[0006] The compound of formula I is preferably prepared starting from α-amino-β-carboalkoxyheterocycles or an α-amino-β-carboxylic acid heterocycle, some of which are commercially available (2 isomers). The methyl thiophene-2-amino-3-carboxylate can be prepared, for example, in accordance with Acta Pharm. Suecica 1968, Vol. 5, p.563, according to S.Gronowitz et al. Other heterocycles can be prepared according to instructions in the literature. The reaction of the α-amino-β-carboalkoxyheterocycles or α-amino-β-carboxylic acid heterocycles with thiophosgene (step 1a in scheme 1) is conveniently carried out in the presence of a base, such as NaOH, KOH, $CaCO_3$, Na_2CO_3, K_2CO_3, $NaHCO_3$, $N(Et)_3$, pyridine, and others, in solvents, such as CH_2Cl_2, $CHCl_3$, ether, tetrahydrofuran and others, possibly in a 2 phase mixture consisting of $CHCl_3$/water or CH_2Cl_2/water, or toluene/water in the temperature range from 0°C to reflux temperature. The resulting isothiocyanates are then converted with primary amines, such as n-butylamine, n-propylamine, isopropylamine, allylamine, propargylamine, cyclopropylamine, and others, in a solvent (ether, tetrahydrofuran, CH_2Cl_2, $CHCl_3$, benzene, toluene, dimethylformamide, dimethylsulfoxide) at 0°C to reflux temperature into the thioureaheterocycles IV (step 2 in scheme 1), which can also be prepared via reaction of the heterocyclic amines II with isothiocyanatoalkanes such as 1-isothiocyanatopropane, 1-isothiocyanatobutane and others in ethanol, n-propanol, n-butanol, dimethylformamide or dimethylsulfoxide as solvents at temperatures between 50°C and reflux temperature (step 1b in scheme 1). The thioureaheterocycles IV, in most cases, cyclise immediately (step 3 in scheme 1). In some cases, the cyclisation is carried out in the presence of stronger bases, such as potassium tert-butylate, sodium hydride or potassium hydride in solvents such as tetrahydrofuran, dimethylfomamide or dimethylsulfoxide in the temperature range from 20°-140°C. The 2-thioxopyrimidin-4-one derivatives are then deprotonised (using bases such as NaOH, NaH, KH, n-BuLi, Na_2CO_3, K_2CO_3 etc.) and are then S-alkylated by the addition of alkylhalides (halo = Br, I) (step 4 in scheme 1). The reaction with methyliodide results in the 2-methylsulfanylpyrimidin-4-one derivative which is

an important intermediate for the synthesis of alkoxy-substituted and aminoalkyl-substituted pyrimidin-4-ones. The replacement of the thiomethyl group (step 5 in scheme 1) with alkoxy is most preferably carried out by reaction with metal alkoxides, such as NaOMe, NaOEt, NaO-propyl, in the corresponding alcohol, tetrahydrofurane or dimethylsulfoxide as solvent in the temperature range from 20°-150°C.

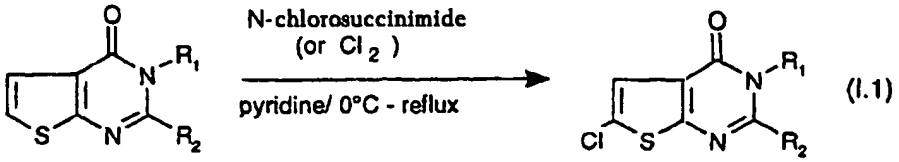
[0007] The above synthesis route is the first disclosure of how to prepare 3H-thieno[2.3-d]-pyrimidin-4-one derivatives within the structural pattern of formula I herein.

[0008] Scheme 3 : Synthesis of special thienopyrimidin-4-ones (special methods for the introduction of halogen into the thiophene ring)

a) Thieno[2.3-d]pyrimidin-4-ones:

[0009]

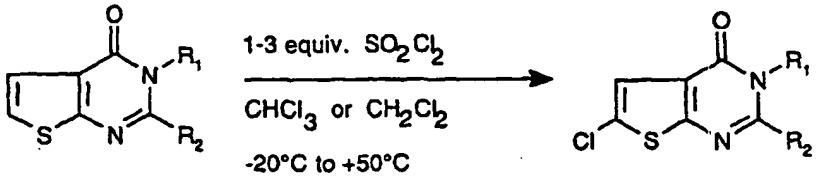
a1)



(I.1)

1-3 molar equivalents of N-chlorosuccinimide (or Cl_2 gas) are used for halogenation. The solvent used is, for example, pyridine in the temperature range from 0°C to reflux. The reaction time is 1 to 24 hours.

a2) "Pure" chlorinating method :



[0010] The described reactions are carried out in per se known manner, e.g. in the presence or absence of a suitable solvent or diluent or of a mixture thereof, if appropriate with cooling, at room temperature or with heating, e.g. in the temperature range from about -20°C to the boiling temperature of the reaction medium, preferably in the range from about -20°C to about +150°C and, if required, in a closed vessel, under pressure, in an inert gas atmosphere and/or under anhydrous conditions.

Illustrative examples of such solvents or diluents are: aromatic, aliphatic and alicyclic hydrocarbons and halogenated hydrocarbons, typically benzene, toluene, xylene, chlorobenzene, bromobenzene, petroleum ether, hexane, cyclohexane, dichloromethane, trichloromethane, dichloroethane or trichloroethane; ethers, typically diethyl ether, tert-butylmethyl ether, tetrahydrofuran or dioxane; ketones, typically acetone or methyl ethylketone; alcohols, typically methanol, ethanol, propanol, butanol, ethylene glycol or glycerol; esters, typically ethyl acetate or butyl acetate; amides, typically N,N-dimethylformamide, N,N-dimethylacetamide, N-methylpyrrolidone or hexamethylphosphoric acid triamide; nitriles, typically acetonitrile; and sulfoxides, typically dimethylsulfoxide. Bases used in excess, such as triethylamine, pyridine, N-methylmorpholine or N,N-diethylaniline, can also be used as solvents or diluents.

Suitable bases are, for example, alkali metal hydroxide or alkaline earth metal hydroxide, alkali metal hydride or alkaline earth metal hydride, alkali metal amide or alkaline earth metal amide, alkali metal alkanolate or alkaline earth metal alkanolate, alkali metal carbonate or alkaline earth metal carbonate, alkali metal dialkylamide or alkaline earth metal dialkylamide, or alkali metal alkylsilylamide or alkaline earth metal alkylsilylamide, alkylamines, atkylenediamines, optionally N-alkylated, optionally unsaturated cycloalkylamines, basic heterocycles, ammonium hydroxides and carbocyclic amines. Examples meriting mention are sodium hydroxide, sodium hydride, sodium amide, sodium methanolate, sodium carbonate, potassiumtert-butanolate, potassium carbonate, lithium diisopropylamide, potassium bis(trimethyl-

silyl)amide, calcium hydride, triethylamine, triethylenediamine, cyclohexylamine, N-cyclohexyl-N,N-dimethylamine, N, N-diethylaniline, pyridine, 4-(N,N-dimethylamino)pyridine, N-methylmorpholine, benzyltrimethylammoniumhydroxide, and 1,8-diazabicyclo[5.4.0]undec-5-ene (DBU).

[0011] Quinazolinone derivatives having fungicidal properties are known from WO-94/26722 or EP-A-276825.

[0012] Surprisingly, it has now been found that the novel compound of formula I has, for practical purposes, a very advantageous spectrum of activities for protecting plants against diseases that are caused by fungi as well as by bacteria and viruses.

The compound of formula I can be used in the agricultural sector and related fields as active ingredient for controlling plant pests. The novel compound is distinguished by excellent activity at low rates of application, by being well tolerated by plants and by being environmentally safe. He has very useful curative, preventive and systemic properties and is used for protecting numerous cultivated plants. The compound of formula I can be used to inhibit or destroy the pests that occur on plants or parts of plants (fruit, blossoms; leaves, stems, tubers, roots) of different crops of useful plants, while at the same time protecting also those parts of the plants that grow later e.g. from phytopathogenic microorganisms.

[0013] It is also possible to use compound of formula I as dressing agents for the treatment of plant propagation material, in particular of seeds (fruit, tubers, grains) and plant cuttings (e.g rice), for the protection against fungal infections as well as against phytopathogenic fungi occurring in the soil.

[0014] The compound I is, for example, effective against the phytopathogenic fungi of the following classes: Fungi imperfecti (e.g. Botrytis, Pyricularia, Helminthosporium, Fusarium, Septoria, Cercospora and Alternaria) and Basidiomycetes (e.g. Rhizoctonia, Hemileia, Puccinia). Additionally, they are also effective against the Ascomycetes classes (e.g. Venturia and Erysiphe, Podosphaera, Monilinia, Uncinula) and of the Oomycetes classes (e.g. Phytophthora, Pythium, Plasmopara). Furthermore, the novel compounds of formula I are effective against phytopathogenic bacteria and viruses (e.g. against Xanthomonas spp, Pseudomonas spp, Erwinia amylovora as well as against the tobacco mosaic virus).

[0015] Within the scope of this invention, target crops to be protected typically comprise the following species of plants: cereal (wheat, barley, rye, oat, rice, maize, sorghum and related species); beet (sugar beet and fodder beet); pomes, drupes and soft fruit (apples, pears, plums, peaches, almonds, cherries, strawberries, raspberries and blackberries); leguminous plants (beans, lentils, peas, soybeans); oil plants (rape, mustard, poppy, olives, sunflowers, coconut, castor oil plants, cocoa beans, groundnuts); cucumber plants (pumpkins, cucumbers, melons); fibre plants (cotton, flax, hemp, jute); citrus fruit (oranges, lemons, grapefruit, mandarins); vegetables (spinache, lettuce, asparagus, cabbages, carrots, onions, tomatoes, potatoes, paprika); lauraceae (avocado, cinnamomum, camphor) or plants such as tobacco, nuts, coffee, eggplants, sugar cane, tea, pepper, vines, hops, bananas and natural rubber plants, as well as ornamentals.

[0016] The compound of formula I is normally used in the form of compositions and can be applied to the crop area or plant to be treated, simultaneously or in succession with further compounds. These further compounds can be e.g. fertilisers or micronutrient donors or other preparations which influence the growth of plants. They can also be selective herbicides as well as insecticides, fungicides, bactericides, nematicides, molluscicides or mixtures of several of these preparations, if desired together with further carriers, surfactants or application promoting adjuvants customarily employed in the art of formulation.

[0017] The compound of formula I can be mixed with other fungicides, resulting in some cases in unexpected synergistic activities.

Mixing components which are particularly preferred are azoles such as propiconazole, difenoconazole, cyproconazole, epoxiconazole, tebuconazole, tetraconazole, fenbuconazole, metconazole, bromuconazole; and also fenpropidine, fenpropimorph, cyprodinile, pyrimethanile, S-methyl benzo-1,2,3-thiadiazole-7-thiocarboxylate; and strobilurines such as azoxystrobine and kresoximemethyl.

[0018] Suitable carriers and adjuvants can be solid or liquid and are substances useful in formulation technology, e. g. natural or regenerated mineral substances solvents, dispersants, wetting agents, tackifiers, thickeners, binders or fertilisers.

[0019] A preferred method of applying a compound of formula I, or an agrochemical composition which contains at least one of said compounds, is foliar application. The frequency of application and the rate of application will depend on the risk of infestation by the corresponding pathogen. However, the compound of formula I can also penetrate the plant through the roots via the soil (systemic action) by drenching the locus of the plant with a liquid formulation, or by applying the compound in solid form to the soil, e.g. in granular form (soil application). In crops of water rice such granulates can be applied to the flooded rice field. The compound of formula I may also be applied to seeds (coating) by impregnating the seeds or tubers either with a liquid formulation of the fungicide or coating them with a solid formulation.

[0020] The compound of formula I is used in unmodified form or, preferably, together with the adjuvants conventionally employed in the art of formulation. To this end they are conveniently formulated in known manner to emulsifiable

concentrates, coatable pastes, directly sprayable or dilutable solutions, dilute emulsions, wettable powders, soluble powders, dusts, granulates, and also encapsulations e.g. in polymeric substances. As with the type of the compositions, the methods of application, such as spraying, atomising, dusting, scattering, coating or pouring, are chosen in accordance with the intended objectives and the prevailing circumstances.

Advantageous rates of application are normally from 5 g to 2 kg of active ingredient (a.i.) per hectare (ha), preferably from 10 g to 1 kg a.i./ha, most preferably from 20 g to 600 g a.i./ha. When used as seed drenching agent, convenient dosages are from 10 mg to 1 g of active substance per kg of seeds.

[0021] The formulation, i.e. the compositions containing the compound of formula I and, if desired, a solid or liquid adjuvant, are prepared in knwon manner, typically by intimately mixing and/or grinding the compound with extenders, e.g. solvents, solid carriers and, optionally, surface active compounds (surfactants).

[0022] Suitable solvents may typically be: aromatic hydrocarbons, preferably the fractions containing 8 to 12 carbon atoms such as xylene mixtures or substituted naphthalenes; phthalates such as dibutyl or dioctyl phthalate; aliphatic hydrocarbons such as cyclohexane or paraffins; alcohols and glycols and their ethers and esters such as ethanol, diethylene glycol, 2-methoxyethanol or 2-ethoxyethanol; ketones such as cyclohexanone; strongly polar solvents such as N-methyl-2-pyrrolidone, dimethyl sulfoxide or dimethyl formamide; as well as vegetable oils or epoxidised vegetable oils such as epoxidised coconut oil or soybean oil; and water.

[0023] The solid carriers typically used for dusts and dispersible powders are usually natural mineral fillers such as calcite, talcum, kaolin, montmorillonite or attapulgite. To improve the physical properties it is also possible to add highly dispersed silicic acid or highly dispersed absorbent polymers. Suitable granulated adsorptive carriers are porous types, including pumice, broken brick, sepiolite or bentonite; and suitable nonsorbent carriers are materials such as calcite or sand. In addition, innumerable pregranulated materials of inorganic or organic origin may be used, typically especially dolomite or pulverised plant residues.

[0024] Depending on the compound of formula I to be formulated, suitable surfactants are nonionic, cationic and/or anionic surfactants having good emulsifying, dispersing and wetting properties. Surfactants will also be understood to include surfactant mixtures.

[0025] Suitable anionic surfactants may be so-called water-soluble soaps as well as water-soluble synthetic surface-active compounds.

[0026] Illustrative examples of nonionic surfactants are nonylphenol polyethoxyethanols, castor oil polyglycol ether, polyadducts of polypropylene and polyethylene oxide, tributylphenoxy polyethoxyethanol, polyethylene glycol and octylphenoxy polyethoxyethanol.

Fatty acid esters of polyoxyethylene sorbitan, such as polyoxyethylene sorbitan trioleate, are also suitable.

[0027] The cationic surfactants are preferably quartemary ammonium salts carrying, as N-substituent, at least one C_8-C_{22} alkyl radical and, as further substituents, optionally halogenated lower alkyl, benzyl or hydroxy-lower alkyl radicals.

Further surfactants customarily employed in the art of formulation are known to the expert or can be found in the relevant literature.

[0028] The agrochemical formulations will usually contain from 0.1 to 99 % by weight, preferably from 0.1 to 95 % by weight, of the compound of formula I, 99.9 to 1 % by weight, preferably 99.8 to 5 % by weight, of a solid or liquid adjuvant, and from 0 to 25 % by weight, preferably from 0.1 to 25 % by weight, of a surfactant.

[0029] Whereas it is preferred to formulate commercial products as concentrates, the end user will normally use dilute formulations.

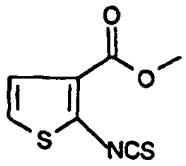
[0030] The compositions may also contain further adjuvants such as stabilisers, antifoams, viscosity regulators, binders or tackifiers as well as fertilisers, micronutrient donors or other formulations for obtaining special effects.

[0031] The following non-limitative Examples illustrate the above-described invention in more detail. Temperatures are given in degrees Celsius. The following abbreviations are used: Et = ethyl; i-propyl = isopropyl; Me = methyl; m.p. = melting point. "NMR" means nuclear magnetic resonance spectrum. MS = mass spectrum. "%" is percent by weight, unless corresponding concentrations are indicated in other units.

Preparation Examples:

Example P-6 Methyl 2-isothiocyanatothiophene-3-carboxylate

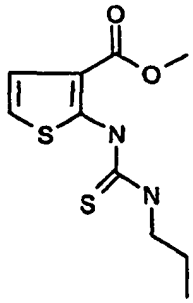
[0032]



[0033] In a sulfonation flask, 50.2 g (0.32 mol) of methyl 2-aminothiophene-3-carboxylate are added to 480 ml of chloroform and 320 ml of water. Then 40.5 9 (0.35 mol) of thiophosgene and 1000 ml of saturated aqueous sodium bicarbonate solution are added simultaneously in 40 minutes under stirring. The stirring continued for 1 h at room temperature and then the organic phase is separated. The water phase is extracted twice with chloroform and the organic phase dried over sodium sulfate. After removal of the chloroform in the water-jet vacuum 61.3 g of a dark oil is obtained, which is further purified by column chromatography over silica gel (eluant: ethyl acetate/hexane=1:5). 41.5 g of methyl 2-isothiocyanatothiophene-3-carboxylate are obtained in the form of a brown powder having a melting point of 63-65°C.

Example P-7 (method 1) Methyl 2-(3-propylthioureido)thiophene-3-carboxylate

[0034]



[cmpd 9.1]

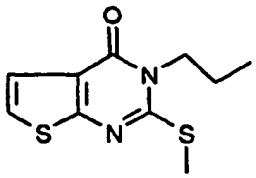
[0035] In a sulfonation flask, 13.5 g (0.023 mol) of n-propylamine are added dropwise to 350 ml of tetrahydrofurane and 41.3 g (0.021 mol) of methyl 2-isothiocyanatothiophene-3-carboxylate, such that the internal temperature does not arise above 40°C. The reaction mixture is then stirred for 4 hours at reflux temperature and then the tetrahydrofurane is removed in a water-jet vacuum. The residue is taken up in ethyl acetate and extracted three times with water. The organic phase is then dried over sodium sulfate and the solvent is removed in the water-jet vacuum, giving the crude product, which is purified by column chromatography over silica gel (eluant: ethyl acetate/hexane=1:3). 32.4 g of methyl 2-(3-propylthioureido)-thiophene-3-carboxylate are obtained in the form of a beige powder having a melting point of 123-126°C.

Example P-7 (method 2) Methyl 2-(3-propylthioureido)thiophene-3-carboxylate

[0036] In a sulfonation flask, 2.02 g (0.02 mol) of 1-isothiocynatopropane are added dropwise to 30 ml dimethylformamide and 3.0 g (0.019 mol) of methyl 2-aminothiophene-3-carboxylate. The reaction mixture is then stirred at 130-135°C for 12 hours and after cooling added to 120 ml of water. The resulting mixture is then extracted three times with ethylacetate and the separeted organic phase dried over sodium sulfate. The solvent is then removed in a water-jet vacuum, giving the crude product as a dark oil, which is purified by column chromatography over silica gel (eluant: tert.butylmethylether/hexane=2:3). 2.0 g of methyl 2-(3-propylthioureido)thiophene-3-carboxylate are obtained in the form of a yellow powder having a melting point of 122-124°C.

Example P-9 2-Methylsulfanyl-3-propyl-3H-thieno[2.3-d]pyrimidin-4-one

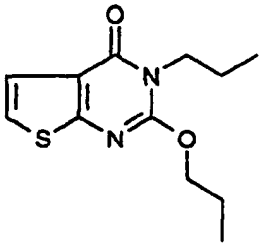
[0037]



[0038] In a sulfonation flask, 2.9 g (0.072 mol) of a ca. 60% sodium hydride dispersion is added to 50 ml of absolute tetrahydrofurane. Then 17.7 g (0.069 mol) of methyl 2-(3-propyithioureido)thiophene-3-carboxylate, dissolved in 100 ml of absolute tetrahydrofurane, are added dropwise, such that the internal temperature remains constant at about 25°C. The mixture is stirred at reflux temperature for 5 hours and after cooling to room temperature 10.9 g (0.077 mol) of methyliodide, dissolved in 10 ml of tetrahydrofurane, are added dropwise. Then the mixture is stirred another 2 hours at reflux temperature. After completion of the reaction, the tetrahydrofurane is removed in tine water-jet vacuum and the residue taken up in ethyl acetate. The organic layer is washed twice with water and then dried over sodium sulfate. After removal of the solvent in the water-jet vacuum, the crude product is obtained, which is purified by digestion in n-hexane. 15.1 g of 2-methylsulfanyl-3-propyl-3H-thieno[2.3-d]pyrimidin-4-one are obtained in the form of a slightly yellowish powder having a melting point of 94-96°C.

Example P-10 2-Propoxy-3-propyl-3H-thieno[2.3-d]pyrimidin-4-one

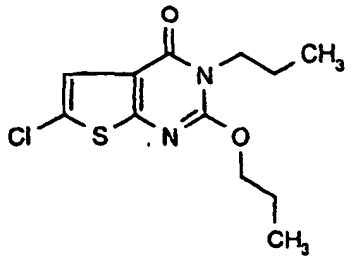
[0039]



[0040] In a sulfonation flask, 12.5 g (0.15 mol) of sodium propylate and 12.0 g (0.05 mol) 2-methylsulfanyl-3-propyl-3H-thieno[2.3-d]pyrimidin-4-one are stirred in 120 ml of absolute n-propanol for 4 hours under nitrogen at reflux temperature. The n-propanol is then removed in a water-jet vacuum and the residue taken up in ethyl acetate/water and the organic phase is extracted twice with water. The organic phase is dried over sodium sulfate and the solvent removed in the water-jet vacuum, giving the crude product, which is then purified by column chromatography over silica gel (eluant: ethyl acetate/hexane=1:3). 6.7 g of 2-propoxy-3-propyl-3H-thieno[2.3-d]pyrimidin-4-one are obtained in the form of a brown resin.

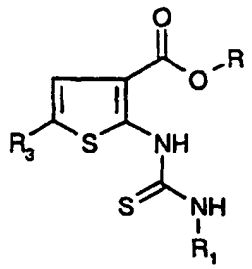
Example P-11 6-Chloro-2-propoxy-3-propyl-3H-thieno[2.3-d]pyrimidin-4-one

[0041]



[0042] In a sulfonation flask, 10.1 g (0.04 mol) of 2-propoxy-3-propyl-3H-thieno[2.3-d]pyrimidin-4-one are added with stirring to 50 ml of absolute pyridine. The internal temperature is then raised to 70-75°C and 9.0 g (0.07 mol) of N-chlorosuccinimid (NCS) are added over about 10 minutes in smallish portions. After stirring for 2 hours at 70-715°C, the pyridine is removed in a water-jet vacuum and the residue taken up in ethyl acetate. After washing twice with cold dilute aqueous hydrochloric acid, drying over sodium sulfate and removing the solvent in a water-jet vacuum, the crude product is obtained. The purification of the crude product is accomplished by column chromatography over silica gel (eluant: ethyl acetate/hexane=1:5), giving 7.8 g 6-chloro-2-propoxy-3-propyl-3H-thieno[2.3-d]pyrimidin-4-one in the form of a white powder having a melting point of 62-65°C.

Table 9



IV'

cmpd No.	R	R_1	R_3	phys. data m.p. °C
9.1	Me	n-propyl	H	123-126

Formulation Examples for compound of formula I

Example F-1.1 to F-1.3: Emulsifiable concentrates

[0043]

Components	F-1.1	F-1.2	F-1.3
compound of formula I	25%	40%	50%

(continued)

Components	F-1.1	F-1.2	F-1.3
calcium dodecylbenzenesulfonate	5%	8%	6%
castor oil polyethylene glycol ether (36 mol ethylenoxy units)	5%	-	-
tributylphenolpolyethylene glycol ether (30 mol ethylenoxy units)	-	12%	4%
cyclohexanone	-	15%	20%
xylene mixture	65%	25%	20%

[0044] Emulsions of any desired concentration can be prepared by diluting such concentrates with water.

Example F-2: Emulsifiable concentrate

[0045]

Components	F-2
compound of formula I	10%
octylphenolpolyethylene glycol ether (4 to 5 mol ethylenoxy units)	3%
calcium dodecylbenzenesulfonate	3%
castor oil polyglycol ether (36 mol ethylenoxy units)	4%
cyclohexanone	30%
xylene mixture	50%

[0046] Emulsions of any desired concentration can be prepared by diluting such concentrates with water.

Examples F-3.1 to F-3.4: Solutions

[0047]

Components	F-3.1	F-3.2	F-3.3	F-3.4
compound of formula I	80%	10%	5%	95%
propylene glycol monomethyl ether	20%	-	-	-
polyethylene glycol (relative molecular mass: 400 atomic mass units)	-	70%	-	-
N-methylpyrrolid-2-one	-	20%	-	-
epoxidised coconut oil	-	-	1%	5%
benzin (boiling range: 160-190°)	-	-	94%	-

[0048] The solutions are suitable for use in the form of microdrops.

Examples F-4.1 to F-4.4: Granulates

[0049]

Components	F-4.1	F-4.2	F-4.3	F-4.4
compound of formula I	5%	10%	8%	21%
kaolin	94%	-	79%	54%
highly dispersed silicic acid	1%	-	13%	7%
attapulgite	-	90%	-	18%

[0050] The novel compound is dissolved in dichloromethane, the solution is sprayed onto the carrier and the solvent is then removed by distillation under vacuum.

Examples F-5.1 and F-5.2: Dusts

[0051]

Components	F-5.1	F-5.2
compound of formula I	2%	5%
highly dispersed silicic acid	1%	5%
talcum	97%	-
kaolin	-	90%

[0052] Ready for use dusts are obtained by intimately mixing all components.

Examples F-6.1 to F-6.3: Wettable powders

[0053]

Components	F-6.1	F-6.2	F-6.3
compound of formula I	25%	50%	75%
sodium lignin sulfonate	5%	5%	-
sodium lauryl sulfate	3%	-	5%
sodium diisobutyinaphthalene sulfonate	-	6%	10%
octylphenolpolyethylene glycol ether (7 to 8 mol ethylenoxy units)	-	2%	-
highly dispersed silicic acid	5%	10%	10%
kaolin	62%	27%	-

[0054] All components are mixed and the mixture is thoroughly ground in a suitable mill to give wettable powders which can be diluted with water to suspensions of any desired concentration.

Biological Examples: Fungidical actions

B-1: Action against Puccinia graminis on wheat

a) Residual-protective action

[0055] 6 days after sowing, wheat plants are sprayed to drip point with an aqueous spray mixture (0.02% active ingredient) prepared from a wettable powder formulation of the test compound and infected 24 hours later with a uredospore suspension of the fungus. After an incubation time of 48 hours (conditions: 95 to 100 % relative humidity at 20°C), the plants are stood at 22°C in a greenhouse. Evaluation of fungal infestation is made 12 days after infection.

b) Systemic action

[0056] Wheat plants are drenched 5 days after sowing with an aqueous spray mixture (0.006% a.i., based on the volume of the soil) prepared from a wettable powder formulation of the test compound. Care is taken that the spray mixture does not come into contact with the growing parts of the plants. After 48 hours, the plants are infected with a uredospore suspension of the fungus. After an incubation period of 48 hours (conditions: 95 to 100 % relative humidity at 20°C), the plants are stood at 22°C in a greenhouse. Evaluation of the fugal infestation is made 12 days after infection. Compound of formula I shows good activity.

Example B-2: Action against Colletotrichum lagenarium on cucumbers

[0057] After a growth period of 2 weeks, cucumber plants are sprayed with an aqueous spray mixture (concentration 0.002%) prepared from a wettable powder formulation of the test compound and infected 2 days later with a spore suspension (1.5×10^5 spores/ml) of the fungus and incubated for 36 hours at 23°C and high humidity. Incubation is then continued at normal humidity and c. 22°C. Evaluation of the fungal infestation is made 8 days after infection.
The compound of formula I shows good activity.

Example B-3: Residual-protective action against Venturia inaequalis on apples

[0058] Apple cuttings with fresh shoots 10 to 20 cm long are sprayed to drip point with a spray mixture (0.02% a.i.) prepared from a wettable powder formulation of the test compound. The plants are infected 24 hours later with a conidia suspensionn of the fungus. The plants are then incubated for 5 days at 90 to 100 % relative humidity and stood in a greenhouse for a further 10 days at 20 to 24°C. Evaluation of the fungal infestation is made 12 days after infection. Compound of formula I shows good activity.

Example B-4: Action against Erysiphe graminis on barley

a) Residual-protective action

[0059] Barley plants about 8 cm in height are sprayed to drip point with a spray mixture (0.02% a.i.) prepared from a wettable powder formulation of the test compound, and the treated plants are dusted with conidia of the fungus 3 to 4 hours later. The infected plants are stood in a greenhouse at 22°C. Evaluation of the fungal infection is made 12 days after infection.

b) Systemic action

[0060] Barley plants about 8 cm in height are drenched with an aqueous spray mixture (0.002% a.i., based on the volume of the soil) prepared from a wettable powder formulation of the test compound. Care is taken that the spray mixture does not come into contact with the growing parts of the plants. The treated plants are dusted 48 hours later with conidia of the fugus. The infected plants are then stood in a greenhouse at 22°C. Evaluation of the fungal infestation is made 12 days after infection.

[0061] Compared with the control plants, infestation of the plants treated with compound of formula I shows no infestation at all.

Example B-5: Action against Podosphaera leucotricha on apple shoots

[0062] Apple cuttings with fresh shoots about 15cm long are sprayed with a spray mixture (0.06% a.i.). The plants are infected 24 hours later with a conidia suspension of the fungus and stood in a climatic chamber at 70% relative humidity and 20°C. Evaluation of the fugal infestation is made 12 days after infection.
Compound of formula I shows 0-5% infestation.

Example B-6: Action against Plasmopara viticola on vines

[0063]

a) Residual-preventive action: Vine cuttings of the Chasselas variety are raised in a greenhouse. At the 10-leaf stage, 3 plants are sprayed with a spray mixture (200 ppm a.i.). After the spray coating has dried, the plants are infected uniformly on the underside of the leaves with a spore suspension of the fungus. The plants are then kept in a humidty chamber for 8 days, after which time marked symptoms of disease are observed on the control plants. The number and size of the infected areas on the untreated plants act as an indicator of the efficacy of the tested compound.

b) Curative action: Vine cuttings of the Chasselas variety are raised in a greenhouse and sprayed at the 10-leaf stage on the underside of the leaves with a spore suspension of Plasmopara viticola. After 24 hours in the humidity chamber, the plants are sprayed with a spray mixture (200 ppm a.i.). The plants are then kept for another 7 days in the humidity chamber. After this time the control plants exhibit symptoms of the disease. The number and size of the infected areas on the untreated plants act as an indicator of the efficacy of the tested compound.

[0064] Compound of formula I shows good efficacy.

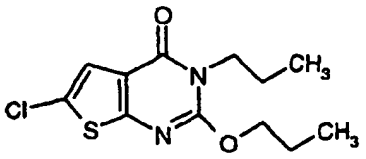
Example B-7: Action against Uncinula necator on vines

[0065] 5 week old vine cuttings are sprayed with a spray mixture (200 ppm a.i.) prepared from a wettable powder formulation of the test compound.The plants are infected 24 hours later by conidias from strongly infested vine leafs that are shaken off over the test plants. The plants are then incubated at 26°C and 60% relative humidity. The evaluation of the fungal infestation is made ca. 14 days after infection.

Compared with the control plants, infestation of the plants treated with compound of formula I shows superior activity (no infestation at all).

Claims

1. The compound of the formula I



I

6-Chloro-2-propoxy-3-propyl-3H-thieno[2.3-d]pyrimidin-4-one.

2. A composition for controlling and preventing pests, wherein the active ingredient is a compound as claimed in claim 1 together with a suitable carrier.

3. Use of a compound of formula I according to claim 1 for protecting plants against infestation by phytopathogenic microorganisms.

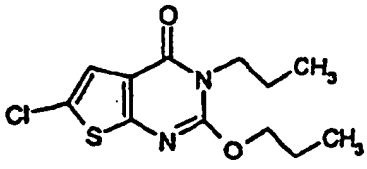
4. A method of controlling or preventing infestation of cultivated plants by phytopathogenic microorganisms by application of a compound of formula I as claimed in claim 1 to plants, to parts thereof or to the locus thereof.

5. A method according to claim 4, wherein the phytopathogenic microorganism is a fungal organism.

6. A method for the preparation of the compound of formula I according to claim 1 which comprises chlorinating 2-propoxy-3-propyl-3H-thieno[2.3-d]-pyridin-4-one with N-chlorosuccinimide or Cl_2 in pyridine or in a mono-, di- or trialkyl-substituted pyridine in the temperature range from 0°C to reflux temperature.

Patentansprüche

1. Verbindung der Formel I



I

nämlich 6-Chlor-2-propoxy-3-propyl-3H-thieno[2.3-d]pyrimidin-4-on.

2. Zusammensetzung für Mittel zur Bekämpfung und Verhütung von Schädlingen, wobei es sich beim Wirkstoff um die Verbindung nach Anspruch 1 zusammen mit einem geeigneten Trägermaterial handelt.

3. Verwendung der Verbindung der Formel I nach Anspruch 1 zum Schutz von Pflanzen gegen der. Befall durch phytopathogene Mikroorganismen.

4. Verfahren zur Bekämpfung oder Verhütung eines Befalls von Kulturpflanzen durch phytopathogene Mikroorganismen, wobei eine Verbindung der Formel I nach Anspruch 1 auf Pflanzenteile oder deren Standort appliziert wird.

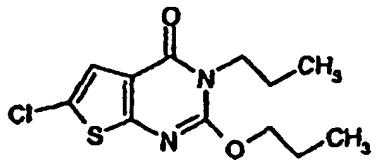
5. Verfahren nach Anspruch 4, wobei es sich beim phytopathogenen Mikroorganismus um einen Pilzorganismus

handelt.

6. Verfahren zur Herstellung der Verbindung der Formel I nach Anspruch 1, umfassend die Chlorierung von 2-Propoxy-3-propyl-3Hthieno[2.3-d]pyrimidin-4-on mit N-Chlorsuccinimid oder Cl_2 in Pyridin oder in einem mono-, di- oder trialkylsubstituierten Pyridin im Temperaturbereich von 0 °C bis zur Rückflusstemperatur.

Revendications

1. composé de formule I



I

6-chloro-2-propoxy-3-propyl-3H-thiéno[2,3-d]-pyrimidin-4-one.

2. composition pour contrôler et prévenir les nuisibles, dans laquelle l'ingrédient actif est un composé selon la revendication 1, ensemble avec un vecteur adéquat.

3. utilisation d'un composé de formule I selon la revendication 1, pour protéger les plantes contre une infestation par des microorganismes phytopathogènes.

4. procédé pour contrôler ou prévenir une infestation de plantes cultivées par des microorganismes phytopathogènes par l'application d'un composé de formule I selon la revendication 1, aux plantes, ou des parties de celles-ci ou aux locus de celles-ci.

5. procédé selon la revendication 4, dans lequel le microorganisme phytopathogène est un organisme fongique.

6. procédé pour la préparation du composé de formule I selon la revendication 1, qui comprend chlorer la 2-propoxy-3-propyl-3H-thiéno[2,3-d]-pyridin-4-one avec la N-chlorosuccinimide ou le Cl_2 dans de la pyridine ou dans une pyridine mono-, di- ou trisubstituée par un alkyle, dans son domaine de température compris entre 0 °C et la température en reflux.